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(54) LAMINATE, FILM OR SHEET AND STRETCH FILM FOR PACKAGING

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a laminate extremely excellent in transparency, flexibility, deformation restoring properties, impact resistance and heat resistance and film, a sheet and a stretch film for packaging comprising the laminate.

SOLUTION: A laminate is constituted of a layer comprising an olefinic resin compsn. (c), which contains 97-5 wt.% of a crystalline olefinic resin (a) and 3-95 wt.% of an amorphous olefinic copolymer (b) obtained by polymerizing at least two kinds of monomer components selected from 3-20C α -olefin and ethylene in the presence of a polymerizing catalyst using a complex of a group IV-VI transition metal having at least one group having a cyclopentadiene type anion skeleton, and an ethylenic resin (d) and a film, a sheet and a stretch film for packaging are composed of this laminate.

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CLAIMS

[Claim(s)]

[Claim 1] Crystalline olefin system resin (a) Under existence of the catalyst for polymerizations which comes to use the transition metal complex of the 4th group of a periodic table who has at least one radical which has 97 - 5 % of the weight and a cyclopentadiene form anion frame - the 6th group, The layer which consists of an olefin system resin constituent (c) containing the amorphous olefin system (copolymer b) 3-95 % of the weight obtained by carrying out the polymerization of at least two kinds of monomer components chosen from the alpha olefin of carbon numbers 3-20, and ethylene, The layered product characterized by consisting of layers which consist of ethylene system resin (d).

[Claim 2] The layered product according to claim 1 which is an amorphous olefin system copolymer which has neither the peak of 1 or more J/g based on fusion of a crystal, nor the peak of 1 or more J/g based on crystallization when an amorphous olefin system copolymer (b) measures with a differential scanning calorimeter (DSC).

[Claim 3] The layered product according to claim 1 which is a propylene- α olefin copolymer with which an amorphous olefin system copolymer (b) consists of a propylene and an α olefin of carbon numbers 4-20.

[Claim 4] The layered product according to claim 1 which is an ethylene-propylene-alpha olefin copolymer with which an amorphous olefin system copolymer (b) consists of ethylene, a propylene, and an alpha olefin of carbon numbers 4-20.

[Claim 5] the mol of the repeat unit to which an amorphous olefin system copolymer (b) is guided from a propylene -- the mol of the repeat unit guided from the alpha olefin of content % (x) and carbon numbers 4-20 -- the layered product according to claim 1 whose P values defined by the following type using content % (y) are 0.3 or more ethylene-propylene-alpha olefin copolymers.

$$P=y/(100-x)$$

[Claim 6] the mol of the repeat unit to which an amorphous olefin system copolymer (b) is guided from a propylene — the mol of the repeat unit guided from the alpha olefin of content % (x) and carbon numbers 4–20 — the layered product according to claim 1 whose P values defined by the following type using content % (y) are 0.5 or more ethylene–propylene–alpha olefin copolymers.

$$P=y/(100-x)$$

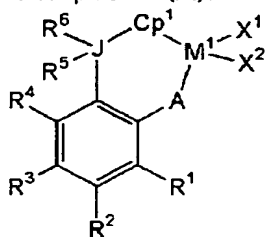
[Claim 7] The layered product according to claim 1 whose molecular weight distribution (Mw/Mn) measured with the gel permeation chromatography (GPC) of an amorphous olefin system copolymer are five or less.

[Claim 8] The layered product according to claim 1 whose limiting viscosity $[\eta]$ by the tetralin solvent in 135 degrees C of an amorphous olefin system copolymer (b) is 0.3 – 10 dl/g.

[Claim 9] The layered product according to claim 1 which is an amorphous olefin system copolymer obtained by carrying out the polymerization of at least two kinds of monomer components chosen from the alpha olefin of carbon numbers 3-20, and ethylene under existence of the catalyst for polymerizations which comes to use the transition metal complex whose amorphous olefin system copolymer (b) is the non-stereospecificity structure of the 4th group of a periodic table who has at least one radical which has a cyclopentadiene form anion frame - the 6th group.

[Claim 10] an amorphous olefin system copolymer (b) -- the following -- (A) and the following -- (B) and/or the following -- the layered product according to claim 1 which is an amorphous olefin system copolymer obtained by carrying out the polymerization of at least two kinds of monomer components chosen from the alpha olefin of carbon numbers 3-20, and ethylene under existence of the catalyst for olefin polymerization which comes to use (C).

Compound (A): The transition metal complex expressed with the following general formula [I]



[I]

(M1 shows the transition-metals atom of the 4th group of the periodic table of an element among a formula, A shows the atom of the 16th group of the periodic table of an element, and J shows the atom of the 14th group of the periodic table of an element.) Cp1 shows the radical which has a cyclopentadiene form anion frame. X1, X2, R1, R2, R3, R4, R5, and R6 show a hydrogen atom, a halogen atom, an alkyl group, an aralkyl radical, an aryl group, a permutation silyl radical, an alkoxy group, an aralkyloxy radical, an aryloxy group, or 2 permutation amino group independently, respectively. It may combine with arbitration and R1, R2, R3, R4, R5, and R6 may form a ring.

(B): one or more sorts of aluminium compound (B1) general formulas chosen from following (B1) - (B3) Organoaluminium compound (B-2) general formula shown by E1aAlZ3-a Annular aluminosilane (B3) general formula which has the structure shown by [-aluminum(E2)-O-] b Linear aluminosilane which has the structure shown by E3[-aluminum(E3)-O-] cAlE32 (however, E1, E2, and E3 are hydrocarbon groups, respectively, and even if all E1, all E2, and all E3 are the same, they may differ.) Z expresses a hydrogen atom or a halogen atom, and even if all Z is the same, it may differ. b expresses two or more integers and c expresses one or more integers for the number with which a is satisfied of $0 < a \leq 3$.

(C): The following (C1) - (C3) one of boron compound (C1) general formulas The boron compound expressed with BQ 1Q2Q3, (C2) General formula The boron compound expressed with G+(BQ 1Q2Q3Q4)-, (C3) General formula The boron compound expressed with (L-H)+(BQ 1Q2Q3Q4)- (However, B is the boron atom of a trivalent valence state, Q1-Q4 are a halogen atom, a hydrocarbon group, a halogenated hydrocarbon radical, a permutation silyl radical, an alkoxy group, or 2 permutation amino group, and even if they are the same, they may differ.) G+ is an inorganic or organic cation, L is a neutral Lewis base, and (L-H)+ is Broensted acid.

[Claim 11] The layered product according to claim 1 whose melt flow rate values of an olefin system resin constituent (c) are 1 - 80g / 10 minutes.

[Claim 12] The layered product according to claim 1 which is crystalline olefin system resin which uses as a principal component the repeat unit to which crystalline olefin system resin (a) is guided from a with a carbon numbers of three or more olefin.

[Claim 13] The layered product according to claim 1 whose crystalline olefin system resin (a) is crystalline propylene system resin.

[Claim 14] The layered product according to claim 1 whose ethylene system resin (d) is at least a kind of ethylene system resin chosen from a low-density-polyethylene and ethylene-alpha olefin copolymer, an ethylene-vinylacetate copolymer, an ethylene-acrylic ester copolymer, and an ethylene-methacrylic ester copolymer.

[Claim 15] The layered product according to claim 1 whose ethylene system resin (d) is the ethylene-vinylacetate copolymer whose content of the repeat unit to which the content of the repeat unit guided from ethylene is guided from vinyl acetate 70 to 95% of the weight is 30 - 5 % of the weight.

[Claim 16] The film or sheet characterized by consisting of a layered product according to claim 1 to 15.

[Claim 17] The stretch film for a package characterized by consisting of a layered product according to claim 1 to 15.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the film which consists of a layered product and this layered product or a sheet, and the stretch film for a package which consists of this layered product. It is related with the suitable layered product for the stretch film for a package which has good transparency, flexibility, deformation recoverability, impact strength-proof, and thermal resistance in the stretch film for a package which puts on a plastic tray etc. and carries out stretch packaging of the food directly in more detail.

[0002]

[Description of the Prior Art] Conventionally, food, such as garden stuff, a fresh fish, fresh meat, and a daily dish, is directly put on a plastic tray etc., and, as for the stretch film for a package which carries out stretch packaging of these with a film, vinyl chloride resin is mainly used. Although based on ethylene system resin, such as low density polyethylene and an ethylene-vinylacetate copolymer, instead of conventional vinyl chloride resin in recent years from an insurance sanitary problem, development is performed actively.

[0003] However, when low density polyethylene etc. was used independently, it could not be satisfied with coincidence of the homogeneity of the film elongation made into the purpose, film rigidity, etc. Moreover, although the ethylene-vinylacetate copolymer film could solve a problem like the above-mentioned low consistency polyethylene film when choosing appropriately a content, molecular weight, etc. of a repeat unit which are guided from vinyl acetate, it had the problem that will be torn when packing food with the case where the angle of a tray is sharp, or a sharp part, and it will be torn.

[0004] For this reason, for example, as shown in JP,2-12187,B or JP,2-18983,B, the laminating of the ethylene-vinylacetate copolymer is carried out to an ethylene-alpha olefin copolymer, and the film for stretch packaging aiming at filling the required engine performance to coincidence is proposed. However, although this film was excellent in mechanical strength, it had the problem that a hole opened on a film with heat-resistant lack in case the film of a tray pars basilaris ossis occipitalis is heat sealed.

[0005] Moreover, the film for stretch packaging which comes to carry out the laminating of the layer which uses an ethylene-vinylacetate copolymer as a principal component to both sides of the layer which uses as a principal component the propylene-alpha olefin random copolymer resin which carried out the polymerization of a kind of alpha olefin chosen from the propylene and the alpha olefin of ethylene and carbon numbers 4-8 to JP,5-147174,A at least is proposed. However, although this film was excellent in thermal resistance, it had the problem that flexibility, deformation recoverability, and impact strength-proof were inferior.

[0006]

[Problem(s) to be Solved by the Invention] The purpose of this invention is to offer the layered product which was extremely excellent in transparency, flexibility, deformation recoverability, impact strength-proof, and thermal resistance, a film or a sheet, and the stretch film for a package.

[0007]

[Means for Solving the Problem] The film or sheet this invention persons excelled [sheet] in transparency, flexibility, deformation recoverability, impact strength-proof, and thermal resistance extremely with the conventional engine performance, And the layer which consists of an olefin system resin constituent which contains the amorphous olefin system copolymer and the crystalline olefin system resin which are obtained using a specific metallocene catalyst as a result of having continued research wholeheartedly about the stretch film for a package, A header and this invention were completed for the film which consists of a layered product which consists of layers which consist of ethylene system resin, and this layered product or a sheet, and the stretch film for a package which consists of this layered product attaining the purpose of this invention.

[0008] Namely, this invention reaches crystalline olefin system resin (a) 97- 5% of the weight. Under existence of the catalyst for polymerizations which comes to use the transition metal complex of the 4th group of a periodic table who has at least one radical which has a cyclopentadiene form anion frame - the 6th group, The layer which consists of an olefin system resin constituent (c) containing the amorphous olefin system (copolymer b) 3-95 % of the weight obtained by carrying out the polymerization of at least two kinds of monomer components chosen from the alpha olefin of carbon numbers 3-20, and ethylene, It is the layered product characterized by consisting of layers which consist of ethylene system resin (d). Moreover, this invention is the film or sheet characterized by consisting of the above-mentioned layered product. Moreover, this invention is a stretch film for a package characterized by consisting of the above-mentioned layered product. Hereafter, this invention is explained to a detail.

[0009]

[Embodiment of the Invention] The amorphous olefin system copolymer (b) used by this invention is an amorphous olefin system copolymer obtained by carrying out the polymerization of at least two kinds of monomer components chosen from the alpha olefin of carbon numbers 3-20, and ethylene under existence of the catalyst for polymerizations which comes to use the transition metal complex of the 4th group of a periodic table who has at least one radical which has a cyclopentadiene form anion frame - the 6th group. As for the amorphous olefin system copolymer (b) used by this invention, it is desirable that the array of a propylene unit and/or an alpha olefin unit side chain is atactic structure. When the orientation of the side chain of the propylene chain in a copolymer is atactic structure as the orientation of the side chain of a propylene unit and/or the alpha olefin unit of carbon numbers 4-20 is atactic structure and the orientation of the side chain of the alpha olefin chain of the carbon numbers 4-20 in a copolymer is atactic structure, the case where the orientation of the side chain of the alpha olefin compound chain of the propylene / carbon numbers 4-20 in a copolymer is atactic structure is shown.

[0010] That the amorphous olefin system copolymer (b) used by this invention is atactic structure For example, the transition metal complex used for the polymerization of an amorphous olefin system copolymer is used. When the polymerization of the gay polypropylene is carried out, the obtained gay polypropylene mm of the propylene methyl carbon which can be determined from a carbon-magnetic-resonance spectrum, F (1) value defined by the following formula using the reinforcement [mm] of each signal which belongs to mr and rr, [mr], and [rr] can check from it or more 40 being the structure which is 55 or less [45 or more] still more preferably 57 or less [43 or more] preferably 60 or less.

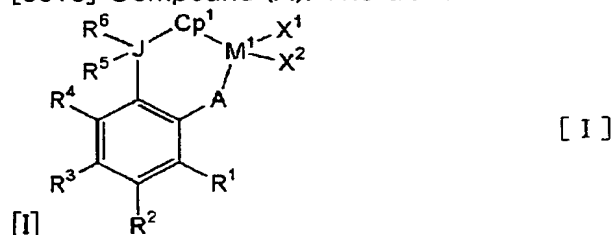
$$F(1) = 100 \times [mr] / ([mm] + [mr] + [rr])$$

[0011] It can check that it is atactic structure because the value equivalent to F (1) called for using the reinforcement of each signal which belongs to mm, mr, and rr(s), such as propylene methyl carbon, branching methylene carbon of an alpha olefin, and branching end methyl carbon of an alpha olefin, similarly from the above backgrounds about the amorphous olefin system copolymer (b) used by this invention is in the above-mentioned range. When an olefin system copolymer is not atactic structure, the flexibility of a layered product and deformation recoverability which are acquired may be inferior. It is presumed to be that to which the amorphous olefin system copolymer used by this invention originates in the array of the propylene in a copolymer and/or an alpha olefin side chain being atactic structure that the

layered product of this invention is excellent in flexibility, deformation recoverability, and transparency. In addition, attribution of mm(s), such as propylene methyl carbon, branching methylene carbon of an alpha olefin, and branching end methyl carbon of an alpha olefin, mr, and rr signal can refer to for example, T.Asakura, Macromolecules, the 24th volume 2334 pages (1991), and the Kinokuniya issue and a new edition giant-molecule analysis handbook (1995).

[0012] As for an amorphous olefin system copolymer (b) since it is above, it is desirable that it is the amorphous olefin system copolymer obtained by carrying out the polymerization of at least two kinds of monomer components chosen from the alpha olefin of carbon numbers 3-20 and ethylene under existence of the catalyst for polymerizations which comes to use the transition metal complex which is the non-stereospecificity structure of the 4th group of a periodic table who has at least one radical which has a cyclopentadiene form anion frame - the 6th group. the transition metal complex which is non-stereospecificity structure -- Cs -- an opposite -- a palm and Cn pair -- it means not having a palm (n being one or more integers). furthermore, an amorphous olefin system copolymer (b) -- the following -- (A) and the following -- (B) and/or the following -- it can manufacture the optimal under existence of the catalyst for olefin polymerization which comes to use (C) by copolymerizing at least two kinds of monomer components chosen from the alpha olefin of carbon numbers 3-20, and ethylene.

[0013] Compound (A): The transition metal complex expressed with the following general formula



(M1 shows the transition-metals atom of the 4th group of the periodic table of an element among a formula, A shows the atom of the 16th group of the periodic table of an element, and J shows the atom of the 14th group of the periodic table of an element.) Cp1 shows the radical which has a cyclopentadiene form anion frame. X1, X2, R1, R2, R3, R4, R5, and R6 show a hydrogen atom, a halogen atom, an alkyl group, an aralkyl radical, an aryl group, a permutation silyl radical, an alkoxy group, an aralkyloxy radical, an aryloxy group, or 2 permutation amino group independently, respectively. It may combine with arbitration and R1, R2, R3, R4, R5, and R6 may form a ring.

[0014] (B): one or more sorts of aluminium compound (B1) general formulas chosen from following (B1) - (B3) Organoaluminium compound (B-2) general formula shown by E1aAlZ3-a Annular aluminosilane (B3) general formula which has the structure shown by {-aluminum(E2)-O-} b Linear aluminosilane which has the structure shown by E3{-aluminum(E3)-O-} cAlE32 (however, E1, E2, and E3 are hydrocarbon groups, respectively, and even if all E1, all E2, and all E3 are the same, they may differ.) Z expresses a hydrogen atom or a halogen atom, and even if all Z is the same, it may differ. b expresses two or more integers and c expresses one or more integers for the number with which a is satisfied of $0 < a \leq 3$.

[0015] (C): The following (C1) - (C3) one of boron compound (C1) general formulas The boron compound expressed with BQ 1Q2Q3, (C2) General formula The boron compound expressed with G+(BQ 1Q2Q3Q4)-, (C3) General formula The boron compound expressed with (L-H)+(BQ 1Q2Q3Q4)- (However, B is the boron atom of a trivalent valence state, Q1-Q4 are a halogen atom, a hydrocarbon group, a halogenated hydrocarbon radical, a permutation silyl radical, an alkoxy group, or 2 permutation amino group, and even if they are the same, they may differ.) G+ is an inorganic or organic cation, L is a neutral Lewis base, and (L-H)+ is Broensted acid.

[0016] Hereafter, the manufacturing method of above-mentioned (A) - (C) is explained in more detail.

(A) In a transition metal complex general formula [I], the transition-metals atom shown by M1 shows the transition-metals element of the 4th group of the periodic table (IUPAC inorganic chemistry nomenclature revised edition 1989) of an element, for example, a titanium atom, a

zirconium atom, a hafnium atom, etc. are raised. They are a titanium atom or a zirconium atom preferably.

[0017] As an atom of the 16th group of the periodic table of the element shown as A in a general formula [I], an oxygen atom, a sulfur atom, a selenium atom, etc. are raised, for example, and it is an oxygen atom preferably.

[0018] As an atom of the 14th group of the periodic table of the element shown as J in a general formula [I], a carbon atom, a silicon atom, a germanium atom, etc. are raised, for example, and they are a carbon atom or a silicon atom preferably.

[0019] As a radical which has the cyclopentadiene form anion frame shown as a substituent Cp1, they are an eta5-(permutation) cyclopentadienyl group, an eta5-(permutation) indenyl group, an eta5-(permutation) fluorenyl group, etc., for example. Specifically An eta5-cyclopentadienyl group, an eta5-methylcyclopentadienyl radical, An eta5-dimethylcyclopentadienyl radical, an eta5-trimethylcyclopentadienyl radical, An eta5-tetramethylcyclopentadienyl radical, an eta5-ethyl cyclopentadienyl group, An eta5-n-propylcyclopentadienyl radical, an eta5-isopropyl cyclopentadienyl group, An eta5-n-butylcyclopentadienyl radical, an eta5-sec-butylcyclopentadienyl radical, An eta5-tert-butylcyclopentadienyl group, an eta5-n-pentyl cyclopentadienyl group, An eta5-neopentylcyclopentadienyl group, an eta5-n-hexyl cyclopentadienyl group, An eta5-n-octylcyclopentadienyl group, an eta5-phenyl cyclopentadienyl group, An eta5-naphthyl cyclopentadienyl group, an eta5-trimethylsilyl cyclopentadienyl group, An eta5-triethyl silyl cyclopentadienyl group, an eta5-tert-butyldimethylsilyl cyclopentadienyl group, An eta5-indenyl group, an eta5-methyl indenyl group, an eta5-dimethyl indenyl group, An eta5-ethyl indenyl group, an eta5-n-propyl indenyl group, an eta5-isopropyl indenyl group,

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TECHNICAL FIELD

[Field of the Invention] This invention relates to the film which consists of a layered product and this layered product or a sheet, and the stretch film for a package which consists of this layered product. It is related with the suitable layered product for the stretch film for a package which has good transparency, flexibility, deformation recoverability, impact strength-proof, and thermal resistance in the stretch film for a package which puts on a plastic tray etc. and carries out stretch packaging of the food directly in more detail.

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PRIOR ART

[Description of the Prior Art] Conventionally, food, such as garden stuff, a fresh fish, fresh meat, and a daily dish, is directly put on a plastic tray etc., and, as for the stretch film for a package which carries out stretch packaging of these with a film, vinyl chloride resin is mainly used. Although based on ethylene system resin, such as low density polyethylene and an ethylene-vinylacetate copolymer, instead of conventional vinyl chloride resin in recent years from an insurance sanitary problem, development is performed actively.

[0003] However, when low density polyethylene etc. was used independently, it could not be satisfied with coincidence of the homogeneity of the film elongation made into the purpose, film rigidity, etc. Moreover, although the ethylene-vinylacetate copolymer film could solve a problem like the above-mentioned low consistency polyethylene film when choosing appropriately a content, molecular weight, etc. of a repeat unit which are guided from vinyl acetate, it had the problem that will be torn when packing food with the case where the angle of a tray is sharp, or a sharp part, and it will be torn.

[0004] For this reason, for example, as shown in JP,2-12187,B or JP,2-18983,B, the laminating of the ethylene-vinylacetate copolymer is carried out to an ethylene-alpha olefin copolymer, and the film for stretch packaging aiming at filling the required engine performance to coincidence is proposed. However, although this film was excellent in mechanical strength, it had the problem that a hole opened on a film with heat-resistant lack in case the film of a tray pars basilaris ossis occipitalis is heat sealed.

[0005] Moreover, the film for stretch packaging which comes to carry out the laminating of the layer which uses an ethylene-vinylacetate copolymer as a principal component to both sides of the layer which uses as a principal component the propylene-alpha olefin random copolymer resin which carried out the polymerization of a kind of alpha olefin chosen from the propylene and the alpha olefin of ethylene and carbon numbers 4-8 to JP,5-147174,A at least is proposed. However, although this film was excellent in thermal resistance, it had the problem that flexibility, deformation recoverability, and impact strength-proof were inferior.

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EFFECT OF THE INVENTION

[Effect of the Invention] As explained in full detail above, according to this invention, the layered product which was extremely excellent in transparency, flexibility, deformation recoverability, impact strength-proof, and thermal resistance can be offered. Moreover, since the layered product of this invention has the above outstanding physical properties, it is suitable for the stretch film for a package. Moreover, the film or sheet of this invention is elasticity, and can be applied to various applications instead of an elasticity vinyl chloride film or a sheet.

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] The purpose of this invention is to offer the layered product which was extremely excellent in transparency, flexibility, deformation recoverability, impact strength-proof, and thermal resistance, a film or a sheet, and the stretch film for a package.

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EXAMPLE

[Example] Hereafter, although an example explains this invention still more concretely, these examples do not limit this invention at all.

[0112] First, the measuring method of the physical-properties value in the following examples and examples of a comparison is explained.

(1) The highest fusion peak temperature (T_m)

After fusing 10mg of samples for 5 minutes at 220 degrees C under nitrogen-gas-atmosphere mind beforehand using a differential scanning calorimeter (DSC by PerkinElmer, Inc.), the temperature was lowered to 40 degrees C at the temperature fall rate for 5-degree-C/. Then, the temperature up was carried out by part for 5-degree-C/, and peak temperature of the maximum peak of the obtained fusion endoergic curve was made into the highest fusion peak temperature (T_m). In addition, the melting point of the indium (In) measured with 5-degree-C programming rate for /using this measuring instrument was 156.6 degrees C. Moreover, about the ethylene-alpha olefin copolymer, about 10mg piece of a trial started from the sheet with a thickness of about 0.5mm produced with a heat press was put into the thump lupane for DSC measurement, preheating was carried out for 5 minutes at 150 degrees C, after lowering the temperature to 40 degrees C and holding for 5 minutes by part for 1-degree-C/, the temperature up was carried out to 150 degrees C the rate for 10-degree-C/, and thermogram was obtained. The ratio of the amount of heat of fusions of the above-mentioned fusion peak to total heat is the value which *(ed) area of the endoergic curve which has the above-mentioned fusion peak in a total endoergic area.

(2) Melt flow rate (MFR)

JIS According to K7210, the ethylene system polymer followed the conditions 4 of Table 1, and it measured by a propylene system polymer following the conditions 14 of Table 1.

(3) Content JIS of a vinyl acetate unit It measured according to K6730.

(4) Transparency (Hayes)

ASTM It measured according to D1003.

[0113] (5) Flexibility (1%SM)

The layered product test piece of the shape of a 120mmx20mm strip of paper which made straight side the direction of taking over (MD) and the direction of taking over, and perpendicular direction (TD) of a layered product was pulled by part for 60mm and speed-of-testing [of 5mm]/between grips, and the stress when being extended 1% was measured.

(6) Deformation recoverability (whenever [deformation recovery])

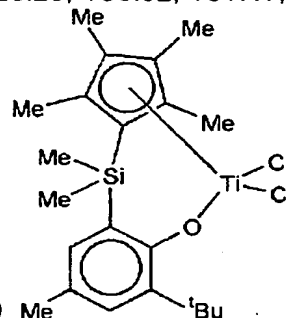
The circular layered product sample 1 with a diameter of 44.45mm was fixed to the layered product fixture 2, and it measured whether using a load cell 4, to the depth 5 predetermined at 100mm rate for /, push ***** and after pulling up at this rate immediately, the push back would disappear completely the pin 3 which set the semi-sphere configuration whose tip is the radius of 6.35mm as the core of a layered product within 30 seconds. The push back considered the maximum pushing depth 5 which disappears completely as whenever [deformation recovery]. Drawing 1 is the top view of the equipment which measures whenever [deformation recovery].

(7) Using Impact strength-proof Oriental energy machine factory film impact circuit tester, 50mmphi made circular the semicircle ball of 15mmphi, and effective test piece area for the

titanium dichloride (3-tert-butyl-5-methyl-2-phenoxy) In the solution which consists of (10.04g), toluene (100ml), and triethylamine (6.30g), at -70 degrees C The 1.63 mols [/.] hexane solution (19.0ml) of n-butyl lithium was dropped, and after that, the temperature up was carried out to the room temperature over 2 hours, and it was further kept warm at the room temperature for 12 hours. Heating reflux was carried out for 10 hours, after dropping the mixture obtained in the top at the toluene solution (50ml) of a titanium tetrachloride (4.82g) and carrying out a temperature up to it to a room temperature over 1 hour after that at 0 degree C under nitrogen-gas-atmosphere mind. A reaction mixture is filtered, a solvent is distilled out of filtrate, a residue is *****ed from a toluene-hexane mixed solvent, and it is dimethylsilyl

(tetramethylcyclopentadienyl) (3-tert-butyl-5-methyl-2-phenoxy) titanium dichloride of an orange columnar crystal. 3.46g was obtained. Yield was 27%. The spectrum data were as follows. 1 H-NMR (CDCl₃) delta 0.57 (s, 6H), 1.41 (s, 9H), 2.15 (s, 6H), 2.34 (s, 6H), 2.38 (s, 3H), 7.15 (s, 1H), 7.18 (s, 1H)

13 C-NMR (CDCl₃) delta 1.25, 14.48, 16.28, 22.47, 31.25, 36.29, 120.23, 130.62, 131.47, 133.86,



135.50, 137.37, 140.82, 142.28, 167.74 mass spectra 458 (Cl, m/e)

[0120] Copolymerization of ethylene, a propylene, and 1-butene was continuously performed using the curing units made from SUS of 100L equipped with the polymerization impeller of [II] amorphous olefin system copolymer (b). That is, the hexane was continuously supplied at the rate of 83L / time amount as a polymerization solvent from the curing-units lower part. On the other hand, polymerization liquid was continuously extracted so that the polymerization liquid in curing units might be set to 100L from the curing-units upper part. Dimethylsilyl (tetramethylcyclopentadienyl) (3-tert-butyl-5-methyl-2-phenoxy) titanium dichloride, triphenylmethyl tetrakis (pentafluorophenyl) borate, and triisobutylaluminum were continuously supplied into curing units from the curing-units lower part an hour an hour as a catalyst by 0.018g / , 0.276g / , and 1.584g/hour in rate, respectively. Moreover, hydrogen performed molecular weight accommodation. Making the jacket attached in the curing-units exterior circulate through cooling water performed the copolymerization reaction at 50 degrees C. A small amount of ethanol is added, the polymerization reaction was stopped, after a monomer and backwashing by water, steam removed the solvent in the polymerization liquid extracted from curing units by underwater [a lot of], the copolymer was taken out in it, and day-and-night reduced pressure drying was carried out to it at 80 degrees C. By the above actuation, the ethylene-propylene-1-butene copolymer ("EPBR" is called hereafter) was obtained by 6.12kg/hour in rate. Conditions and a result are shown in Table 1.

[0121] It carried out on the same conditions as the example 1 of reference except having changed the amount of example of reference 2 catalyst, and the hydrogen used. Conditions and a result are shown in Table 1. In addition, the measuring method of the content of the propylene unit in EPBR obtained in the example 1 of reference and the example 2 of reference and 1-butene unit is as follows.

[0122] (a) The heat press was carried out, respectively and the mixture of various mixed ratios of the creation propylene homopolymer of a calibration curve and an ethylene-1-butene copolymer was fabricated with a thickness of 0.05mm in the shape of a film. It asked for the absorbance of a propylene unit origin peak (wave number 1150cm⁻¹) and 1-butene unit origin peak (wave number 770cm⁻¹) using the infrared spectrometer, and the propylene and 1-butene unit content in this mixture were plotted to this absorbance. In quest of the regression line, it

considered as the calibration curve from these plots. In addition, after the mixture of a propylene homopolymer and an ethylene-1-butene copolymer dissolved both in toluene, the precipitate which added the methanol and was obtained was dried and used for it.

[0123] (b) The heat press of the measurement olefine copolymer of the content of a propylene unit and 1-butene unit was carried out, and it fabricated with a thickness of 0.05mm in the shape of a film, and, subsequently asked for the absorbance of a propylene unit origin peak and 1-butene unit origin peak using infrared spectrometer, and the content of the propylene unit in an olefine copolymer and 1-butene unit was computed from the calibration curve acquired from the above-mentioned approach.

[0124] Copolymerization of a propylene and 1-butene was continuously performed using the curing units made from SUS of 100L equipped with example of reference 3 impeller. That is, the hexane was continuously supplied at the rate of 83L / time amount as a polymerization solvent from the curing-units lower part. On the other hand, polymerization liquid was continuously extracted so that the polymerization liquid in curing units might be set to 100L from the curing-units upper part. Dimethylsilyl (tetramethylcyclopentadienyl) (3-tert-butyl-5-methyl-2-phenoxy) titanium dichloride, triphenylmethyl tetrakis (pentafluorophenyl) borate, and triisobutylaluminum were continuously supplied into curing units from the curing-units lower part an hour an hour as a catalyst by 0.068g / , 2.067g / , and 3.964g/hour in rate, respectively. Moreover, hydrogen performed molecular weight accommodation. Making the jacket attached in the curing-units exterior circulate through cooling water performed the copolymerization reaction at 50 degrees C. A small amount of ethanol is added, the polymerization reaction was stopped, after a demonomer and backwashing by water, steam removed the solvent in the polymerization liquid extracted from curing units by underwater [a lot of], the copolymer was taken out in it, and day-and-night reduced pressure drying was carried out to it at 80 degrees C. By the above actuation, the propylene-1-butene copolymer ("PBR" is called hereafter) was obtained by 5.60kg/hour in rate. Conditions and a result are shown in Table 1. In addition, the measuring method of 1-butene content in obtained PBR is as follows.

(a) The heat press was carried out, respectively and the creation propylene of a calibration curve and the mixture of various mixed ratios of 1-butene homopolymer were fabricated with a thickness of 0.05mm in the shape of a film. It asked for the extinction quotient of a propylene unit origin peak (wave number 1150cm⁻¹) and 1-butene unit origin peak (wave number 770cm⁻¹) using the infrared spectrometer, and the content of 1-butene unit in this mixture was plotted to this extinction quotient. In quest of the regression line, it considered as the calibration curve from these plots. In addition, after the mixture of the independent copolymer of a propylene and 1-butene dissolved both in toluene, the precipitate which added the methanol and was obtained was dried and used for it.

[0125] (b) The heat press of the measurement propylene-1-butene copolymer of the content of 1-butene unit was carried out, and it fabricated with a thickness of 0.05mm in the shape of a film, and, subsequently asked for the extinction quotient of a propylene unit origin peak and 1-butene unit origin peak using infrared spectrometer, and the content of 1-butene unit in a propylene-1-butene copolymer was computed from the calibration curve acquired from the above-mentioned approach. differential scanning calorimeter (DSC) measurement -- a differential scanning calorimeter (DSCby SEIKO electronic industry company220C) -- using -- a temperature up and constant temperature -- all of a process measured the rate for 10-degree-C/.

[0126] Measurement of limiting viscosity [eta] was performed using the ubellohde's viscosimeter in 135-degree-C tetralin. The sample dissolved 300mg in 100ml xylene, and adjusted the 3mg [/ml] solution. Furthermore, the solution concerned was diluted to 1/2, 1/3, and 1/5, and each was measured in the 135 degrees C (**0.1 degrees C) constant temperature bath. By each concentration, it measured repeatedly 3 times, and the acquired value was averaged and used.

[0127] molecular weight distribution -- a gel permeation chromatograph (GPC) -- it carried out by law (the product made from Waters, 150 C/GPC equipment). Elution temperature is 140 degrees C and the column used is Sodex by Showa Denko K.K. Packed ColumnA-80M and a molecular-weight reference material used polystyrene (the TOSOH CORP. make, molecular

weight 68-8,400,000). The obtained polystyrene equivalent weight average molecular weight (Mw), number average molecular weight (Mn), and also this ratio (Mw/Mn) were made into molecular weight distribution. The measurement sample made about 5mg polymer the dissolution and the concentration of about 1mg/ml at 5ml o-dichlorobenzene. Injection of the 400microl of the obtained sample solution was carried out. The elution solvent rate of flow was considered as a part for 1.0ml/, and the refractive index detector detected it.

[0128]

[Table 1]

項 目	単位	参考例 1	参考例 2	参考例 3
重合温度	℃	50	50	50
エチレン	kg/h	2	2	0
プロピレン	kg/h	8.3	8.3	13.0
1-ブテン	kg/h	12.7	12.7	3.3
* 1 (a)	g/h	1.584	1.584	3.964
* 2 (b)	g/h	0.276	0.138	2.067
* 3 (c)	g/h	0.018	0.009	0.068
エチレン含有量	mol%	11	15	0
プロピレン含有量	mol%	40	37	78
1-ブテン含有量	mol%	49	48	22
結晶融点	℃	なし	なし	なし
結晶融解熱量	mj/mg	なし	なし	なし
結晶化温度	℃	なし	なし	なし
結晶化熱量	mj/mg	なし	なし	なし
極限粘度 [η]	dl/g	0.94	1.04	0.84
G P C Mw/Mn	—	2.2	2.3	1.8

[0129] * 1(a): -- triisobutylaluminum *2 -- (b):triphenylmethyl tetrakis (pentafluorophenyl) borate *3(c):dimethylsilyl (tetramethylcyclopentadienyl) (3-tert-butyl-5-methyl-2-phenoxy) titanium dichloride [0130] 98 % of the weight (the content of Eve Tait H2081 by Sumitomo Chemical Co., Ltd., 10 MFR(190 degrees C) =2g /, and minutes, and a vinyl acetate unit = 15.8 % of the weight) of example 1 [preparation of surface layer resin constituent] ethylene-vinylacetate copolymers and the resin constituent which carried out melting kneading and prepared 2 % of the weight (Marubishi Yuka Kogyo STO- 405) of antifoggers with the Banbury mixer were used as a resin constituent which constitutes a surface layer.

[preparation of a middle class resin constituent] -- as a crystalline olefin system polymer -- a propylene-ethylene random copolymer (no BUREN S131 by Sumitomo Chemical Co., Ltd., and Tm= -- 135 degrees C) 5-% of the weight 49 % of the weight, The content of MFR(230 degrees C) =1.0g /, 10 minutes, content =95% of the weight of a propylene unit, and an ethylene unit = 49 % of the weight of EPBR(s) obtained in the example 1 of reference as an amorphous olefin system copolymer, after [and] carrying out melting kneading of the 2 % of the weight (Marubishi Yuka Kogyo STO- 405) of the antifoggers with a Banbury mixer -- further -- the resin constituent which carried out melting kneading and which was prepared with the 1 shaft extruder was used as a resin constituent which constitutes an interlayer.

The ethylene-vinylacetate copolymer constituent [manufacture of a layered product] Obtained to a surface layer The two-sort three-layer layered product which has in an interlayer a resin constituent containing a propylene-ethylene random copolymer and EPBR obtained in the example 1 of reference is supplied to the Placo three-Layer inflation film processing machine. Thickness percentage manufactured the layered product with a total thickness of 13

micrometers 38% / 24% / 38% in order of the surface layer / interlayer / surface layer by processing it by the dice temperature of 200 degrees C, and the blow ratio 4.5. Many characteristic values of the obtained layered product are shown in Table 2.

[0131] Thickness percentage manufactured the layered product with a total thickness of 13 micrometers 38% / 24% / 38% in order of the surface layer / interlayer / surface layer like the example 1 except having used EPBR obtained in the example 2 of reference as example 2 amorphous olefin system resin. Many characteristic values of the obtained layered product are shown in Table 2.

[0132] Thickness percentage manufactured the layered product with a total thickness of 13 micrometers 38% / 24% / 38% in order of the surface layer / interlayer / surface layer like the example 1 except having used PBR obtained in the example 3 of reference as example 3 amorphous olefin system resin. Many characteristic values of the obtained layered product are shown in Table 2.

[0133] Thickness percentage manufactured the layered product with a total thickness of 13 micrometers 38% / 24% / 38% like the example 1 in order of a surface layer / middle class / surface layer except having considered as the propylene-ethylene random copolymer which used the example of comparison 1 [manufacture of layered product] middle class resin constituent in the example 1. Many characteristic values of the obtained layered product are shown in Table 2.

[0134]

[Table 2]

項 目	実施例 1	実施例 2	実施例 3	比較例 1
歪 率 (%)	1. 2	1. 2	1. 1	1. 5
1 % SM MD (kg/cm ²)	5 5 0	4 3 0	5 9 0	1 4 4 0
同上 TD (kg/cm ²)	5 2 0	3 8 0	5 9 0	1 4 6 0
変形回復度 (mm)	1 4	1 5	1 4	8
耐衝撃強度 (kg・cm/mm)	2 1 0	2 2 0	2 0 0	1 8 0
耐熱温度 (°C)	1 1 0	1 1 0	1 1 0	1 1 0

[Translation done.]

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1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. **** shows the word which can not be translated.
3. In the drawings, any words are not translated.

DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] It is the top view of the equipment which measures whenever [deformation recovery].

[Description of Notations]

1 [... A load cell, 5 / ... Pushing depth] ... A layered product sample, 2 ... A layered product fixture, 3 ... A pin, 4

[Translation done.]

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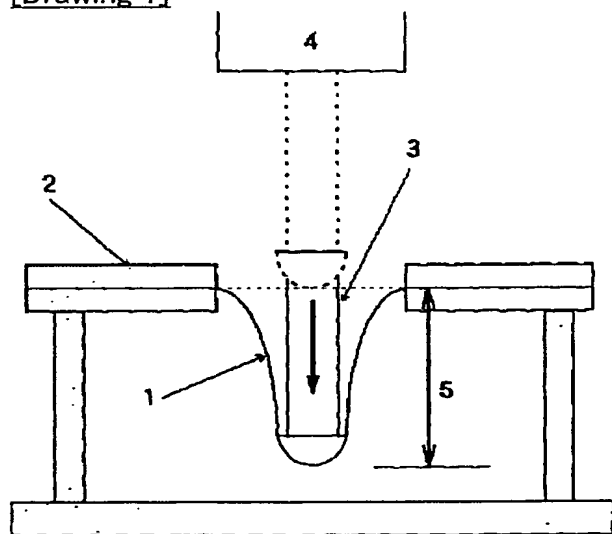
1.This document has been translated by computer. So the translation may not reflect the original precisely.

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DRAWINGS

[Drawing 1]



[Translation done.]